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Abstract

Cleaning products are among the most widely used consumer products. The associated risks should be better understood. The health risk assessment (HRA) approach was applied to household uses of cleaning products, with nineteen products of various types and formats tested under typical indoor environmental parameters in an experimental house. The targeted substances included volatile organic compounds (VOCs) and carbonyl compounds. The generic "Common Use" and "Reasonable Worst-Case" scenarios under consideration were based on full cleaning sessions. These sessions were elaborated from data available in the technical and scientific literature, combined with stakeholder participation. The Common Use scenario included a 1 1/2-hour cleaning session once per week, followed by manual ventilation; the Reasonable Worst-Case scenario included a 4-hour session twice per week without manual ventilation. No situation of concern was found regarding chronic inhalation exposures associated with Common Use. For the Reasonable Worst-Case scenario, the assessed chronic inhalation risks were low. Assessed acute inhalation exposures (1-hour exposures) could exceed the selected health values, mainly for acrolein (exposures up to 12 µg/m^3) and formaldehyde (exposures up to ~140 µg/m^3). Associated first observed effects could include nasal, throat, and eye irritation. These results suggest that the highest exposures should be reduced and, to this end, that the emissions of the most emissive products should be reduced. Since the identified priority substances of concern are not specific to cleaning product emissions, multisource cumulative exposures are expected with the use of other consumer products, e.g., paints, incense, scented candles, furniture, and fragrance diffusers.

Keywords

Consumer products, indoor air quality, generic exposure scenarios, stakeholder participation, household cleaning sessions, inhalation exposures.

Abbreviations

AIC, average inhaled concentration; ARR, acute risk ratio; HAC_{max}, maximum 1-hour average concentration; IER, individual excess risk; TER, threshold effect ratio; TRV_T, threshold toxicity reference value; TRV_{NT}, nonthreshold toxicity reference value; TRV_{acute}, acute toxicity reference value; VOCs, volatile organic compounds

1 Introduction

In temperate climates, most people spend nearly 90% of their time in indoor environments, mainly at home. Consequently, indoor air quality is a topic of major importance to public health [1-3].

Cleaning products are widely used household consumer products [4, 5] that are intended to make cleaning the house easier. They constitute a specific indoor emissions source of air pollutants, e.g., carbonyl compounds, aerosols, and terpenes [2, 6-10], and they are a subject of concern [11-15], especially for children [16, 17] due to the specific metabolism and vulnerabilities associated with their developing bodies [18, 19]. The associated risks should be better understood [20-22]. In particular, few health risk assessments (HRAs) have been conducted in the scientific literature; they have focused on a limited selection of products tested in emissions test chambers and on modeling a few target substances [23-25]. Furthermore, available data about household uses of cleaning products have mainly been provided about generic types of products, i.e., wide thematic categories of products, e.g., all-purpose cleaners, bathroom cleaners, window cleaners, etc. These types of products have been considered individually; i.e., the available data are specific to one particular type of product, regardless of the potential uses of other types of products, e.g., the use frequency of a window cleaner with no information about the potential use of an all-purpose cleaner in the same cleaning session [10, 26-31]. In other words, the available data do not describe the uses of multiple products in a cleaning session (“co-uses”) and how these uses vary among the different rooms of a private dwelling. Indeed, characterizing the co-uses of full, multiroom and multiproduct household cleaning sessions constitutes a challenge [5, 27].

This study aimed to contribute to a better characterization of the inhalation exposures and risks associated with household uses of cleaning products: an HRA was conducted for nineteen cleaning products of various types and formats, tested under realistic indoor conditions (i.e., under typical indoor environmental parameters). In particular, this HRA aimed to identify the emitted substances of greatest concern and to assess the associated health issues.

This study is based on the measurement data produced in the ADOQ (Domestic Activities and Indoor Air Quality) project [32].

2 Materials and Methods

2.1 Experiments under realistic conditions

Nineteen cleaning products were selected, aiming èto obtain various types (e.g., floor cleaners, all-purpose cleaners, bathroom cleaners, window cleaners) and formats (e.g., liquids, sprays, wipes, creams) among those commonly used in France [33].

Emissions into indoor air from the selected products were tested in the “Mechanized House for Advanced Research on Indoor Air” (MARIA; Marne-la-Vallée, France) [34]. Measurements were performed in a 32-m^3 bedroom, an 18-m^3 bathroom and a 27-m^3 kitchen. These rooms are empty of additional furniture, and the finishing coatings are minimal: raw concrete floor, painted plasterboard walls, and painted concrete ceiling.

All of the experiments were conducted under realistic conditions, especially regarding the air exchange rate. The indoor air conditions of the rooms, e.g., temperature and relative humidity, were monitored continuously. The ventilation of each room was controlled.

A detailed physicochemical characterization of the emissions was performed [9, 32, 35, 36] using online instrumentation (including Aerosol Mass Spectrometer Compact Time-of-Flight (AMS-c-TOF); specific analyzers (compact-GC-FID for C2-C6 and C6-C12; ozone; nitrogen oxides); FID chromatograph; and Scanning Mobility Particle Sizer (SMPS)) and off-line chemical analysis for gaseous phase (DNPH cartridges and Tenax TA adsorbent tubes) and particulate phase (filters). This instrumentation allowed us to characterize both primary emitted substances and secondary substances formed during and after use and to monitor emitted fine particles of concern (e.g., submicron and ultrafine particles, including ozone-induced secondary particles).

Volatile organic compounds (VOCs) and carbonyl compounds were sampled and analyzed according to ISO 16000-6 [37] and 16000-3 [38], respectively. To search for limonene oxidation tracers, a specific analytical approach was employed; i.e., molecular composition was investigated using derivatization prior to thermal desorption coupled with gas chromatography and mass spectrometry (TD-GC-MS) analysis.

Two sets of experiments were performed: the first in summertime and the second in wintertime. Following the recommendations of the manufacturers, the selected cleaning products were applied with predetermined procedures (see details in Supplementary Material), including specific durations of use.

After checking the background level of pollutants in the room, particulates and volatile gaseous compounds were measured during the five 30-min periods following the beginning of use (i.e., from 0 to 30 min, from 30 to 60 min, from 60 to 90 min, from 90 to 120 min, and from 120 to 150 min, respectively). The first "0–30 min" measurement period included the use phase of the product. Air samples were obtained close to the user, approximately at the height of airways (1.50 m) in a zone associated with efficient air mixing.

After reviewing the scientific literature [7, 10, 15, 20, 23-25, 39] and pretesting in emissions test chambers, the selected targeted substances included VOCs, such as benzene, toluene, ethylbenzene, styrene, xylenes, and naphthalene; carbonyl compounds, such as formaldehyde, acetaldehyde, acetone, propionaldehyde, benzaldehyde and acrolein; and terpenes, such as d-limonene and alpha-pinene. Semivolatile organic compounds (SVOCs) were not in the scope of this HRA.

2.2 Elaboration of generic exposure scenarios

2.2.1 Common Use and Reasonable Worst-Case scenarios

Household exposures to cleaning product emissions can vary with multiple factors, e.g., product uses (which can vary with age, gender, sociodemographic status, geographical area, etc.), ventilation practices, the environmental characteristics of the exposed persons (e.g., ventilation and volume of the considered rooms), and times spent in each considered room (which can vary with the types of inhabitants, e.g., children, students, housekeepers, retired people, salaried adults). Regarding the difficulty of managing the complexity of such multicriterion variability, the choice was made to base the HRA, as a first approach, on two types of generic exposure scenarios, i.e., the Common Use scenario and the Reasonable Worst-Case scenario. This choice allowed us to study two substantially different levels of exposure as a simple way to characterize the actual variability. The Common Use scenario aimed to characterize a user whose practices and environmental characteristics (volume of the room, air change rate, etc.) are common, being approximately the average of the observed practices and characteristics according to French and European surveys [4, 40, 41]. The Reasonable Worst-Case scenario aimed to characterize a user whose practices and environmental characteristics reasonably increase the average exposure (reasonably conservative choices, according to French and European surveys).

Each scenario was defined by several components: type of room to be cleaned, volume (m^3) and daily presence (h/d) for each considered room; ventilation (air change rate expressed in changes/h); duration of exposure in a lifetime (years); potential manual ventilation of the room (i.e., a user opens a window to temporarily increase the natural ventilation level) during or after the use of cleaning products; cleaning session, i.e., types of products used in each room; and the associated frequencies and durations of use. To be integrated with the Common Use scenario and the Reasonable Worst-Case scenario (Table 1; Table 2), a Common Use session and a Reasonably Elevated Use session were elaborated based on the same logics.

2.2.2 Cleaning sessions elaborated with data available in the literature

A review of the technical and scientific literature was conducted to collect and analyze available data about household uses of cleaning products. These data (e.g., quantities used, frequencies and durations of use) are mainly provided by wide thematic categories of products considered individually; i.e., the available data are specific to one category potentially used in a cleaning session, regardless of the potential uses of other categories, e.g., use frequency of a floor cleaner with no information about the potential use of an all-purpose cleaner in the same cleaning session [10, 26-31]. Although some studies have provided data on co-uses (i.e., uses of different products within the same cleaning session) [40, 42, 43], they did not describe how the studied co-uses varied with the considered rooms and influenced the use levels of each type of product; e.g., an all-purpose cleaner user may tend to result in use of fewer specific-purpose products (floor cleaner, window cleaner, bathroom cleaner, etc.). In particular, the available data did not allow us to characterize detailed co-uses of full, multiroom and multiproduct household cleaning sessions [5, 27].

Aiming to come closer to the observed household practices, the exposure assessment of this HRA was based on two elaborated full cleaning sessions: a Common Use session, which aims to describe a user whose practices are common and which are in the average of the observed practices; and a Reasonably Elevated Use session, which aims to describe a user whose practices reasonably increase common levels of use. Each session consists of several components: rooms to be cleaned; for each room considered, types of product used; and for each type of product in each room, the frequency and duration of use.

First, a proposal for the two considered sessions was elaborated based on best compromises between data available in the technical and scientific literature [4, 29, 40, 42, 44-46]. For each session, the selected values of each component aimed to obtain sessions that were reasonably coherent with as many sources of information as possible. Typically, the components of the Common Use session and of the Reasonably Elevated Use session were chosen from the approximately 50^{th} and 90^{th} percentiles of the collected data, respectively. In the case of substantially conflicting data or data with substantial variability, the preference choice targeted the most detailed and recent data produced by public funding and, if necessary, characterizing French uses.

2.2.3 Stakeholder participation

The elaborated cleaning sessions were sent for comments to a selection of stakeholders, i.e., persons or organizations that, regarding their activity, are interested in the results of this HRA. The selected stakeholders were from various backgrounds, e.g., companies providing home cleaning services, indoor environment medical consultants, environmental health associations, environmental health public organizations, family associations, cleaning product manufacturers, consumer associations, and private housekeepers. The objectives of this consultation were: (i) to identify potential inconsistencies in the elaborated sessions; and (ii) in the absence of statistically representative data, to collect feedback from different stakeholder profiles to contextualize the values selected for the cleaning sessions of this study. For example, each stakeholder profile could provide a different set of responses to the following questions: "*Is a type of product missing in the considered cleaning sessions? For each considered type of product in each considered room, is the selected average duration appropriate? What do you think about the total length of the considered sessions?*" These different feedbacks are all elements of knowledge, which could support a proactive critical analysis.

2.3 Exposure assessment

This HRA aimed to contribute to a better characterization of inhalation exposures. Other exposure routes (e.g., ingestion and dermal contact) were not within the scope of the work.

A house with classic types of rooms was considered for this HRA, with the following characteristics: kitchen (volume: 30 m^3; daily presence: 2.75 h/d), bedroom (45 m^3; 8.5 h/d), bathroom (24 m^3; 1 h/d), living/dining room (90 m^3; 5.75 h/d), restroom (5 m^3; 0.5 h/d), entrance hall (12 m^3; 0.5 h/d) and storeroom (10 m^3; 0.5 h/d). An air change rate of 0.35/h was retained for the entire house. These characteristics are consistent with those used for other European studies [10, 47], including the HRA of the European EPHECT project (considered population groups: housekeepers and retired people).

Sorption/desorption phenomena and migration within the house were not quantified, assuming that the associated variability is much lower than the uncertainties associated with the other assumptions of the HRA.

The concentrations measured under realistic conditions were extrapolated to daily average concentrations based on the following hypotheses: 10 minutes of manual ventilation after use is supposed to renew the air of the room (Common Use scenario) [4, 48]; otherwise, concentrations in the room are supposed to be influenced only by the selected air change rate (Reasonable Worst-Case scenario). For each elaborated scenario (Table 1; Table 2) and each emitted substance, these hypotheses were used to derive concentrations at each time of the day in each room. Then, daily average concentrations were calculated in each room.

From these daily average concentrations, average inhaled concentrations (AICs) were calculated by applying the components of each generic exposure scenario, e.g., daily durations of use and daily presence in each room. For each emitted substance, the AICs of each room were summed to obtain an AIC characterizing an average daily presence in the whole house under consideration (AIC_{house}).

These calculated AIC_{house} values characterized chronic inhalation exposures.

Acute exposures were also assessed. In general, this type of exposure corresponds to periods that could vary from one hour to a few days. Considering the typical household uses of cleaning products, an exposure of one hour was retained for the HRA. Therefore, acute exposures were characterized by maximum 1-hour average concentrations (HAC_{max}), calculated with the two highest maximum half-hour exposures (minimum considered daily presence in a room), among all of the considered rooms.

2.4 Selection of toxicity values

For both chronic effects and acute effects, a collection and a choice of toxicological reference values (TRVs) were made from the TRVs present in usual reference databases -- including those from the United States Environmental Protection Agency (US EPA), World Health Organization (WHO), Agency for Toxic Substances and Disease Registry (ATSDR), French Agency for Food, Environmental and Occupational Health & Safety (ANSES), Health Canada, Dutch National Institute for Public Health and the Environment Ministry of Health (RIVM) and California Office of Environmental Health Hazard Assessment (OEHHA) -- for each tested substance. This choice was made for threshold effects (TRV_T), nonthreshold effects (TRV_{NT}) and acute effects (TRV_{acute}). Since HAC_{max} was selected to characterize acute exposures, the TRVs associated with a 1-hour exposure were chosen preferentially.

When no TRV was available for chronic exposures, the derived European Lowest Concentration of Interest (EU-LCI) [49], elaborated with a method similar to that used for TRVs, was considered additional information.

2.5 Health risk assessment

Based on the exposures calculated with the concentrations measured under realistic conditions and with the elaborated generic exposure scenarios, each including a full cleaning session, an HRA was conducted [50, 51].

The scope of this HRA focused on the inhalation route of exposure. Chronic threshold and nonthreshold risks, as well as acute risks, were quantified by the following risk indicators: threshold effect ratio (TER), individual excess risk (IER) and acute risk ratio (ARR).

$$\boldsymbol{TER = \frac{AIC_{house}}{TRV_T}} \qquad \textbf{(1)}$$

$$\boldsymbol{IER = \frac{AIC_{house} \times TRV_{NT} \times Years\ of\ exposure}{Lifetime\ of\ 70\ years}} \qquad \textbf{(2)}$$

$$\boldsymbol{ARR = \frac{HAC_{max}}{TRV_{acute}}} \qquad \textbf{(3)}$$

where TER is the threshold effect ratio, AIC is the average inhaled concentration, TRV_T is the threshold toxicity reference value, IER is the individual excess risk, TRV_{NT} is the nonthreshold toxicity reference value, ARR is the acute risk ratio, HAC_{max} is the maximum 1-hour average concentration, and TRV_{acute} is the acute toxicity reference value.

3 Results

3.1 Elaboration of generic exposure scenarios

Stakeholder participation included a total of 32 requests. Twenty-three responses were obtained, representing a return rate of 72%. Overall, except for two people, all of the participants considered that the proposed sessions (Table S2) corresponded to the correct orders of magnitude of total time for a "Common Use" session and for a "Reasonably Elevated Use" session. Based on the detailed answers, modifications were retained when suggested by more than half the participants, e.g.: (i) the daily duration of use for dishwashing products was reduced from 30 minutes to 10 minutes in the Common Use session since more than two-thirds of the participants indicated that the use of a dishwasher was the most common situation at the time; and (ii) the use of dusting products was no longer considered since more than two-thirds of the participants indicated that the use of microfiber cloths is now more widespread.

No modifications were applied to the Reasonably Elevated Use session. Overall, opinions about this session did not show any marked trends, with one exception: the majority of the participants spontaneously indicated that the total time should be maintained, arguing that (i) such practices are plausible for a substantial part of the general population and that (ii) the exposure of those who believe in the usefulness of a high level of product use should not be underestimated. This HRA was applied based on the updated cleaning sessions (Table 1).

For other components of the generic exposure scenarios, e.g., air change rate, volumes and daily presences in the rooms selected in each scenario, the values retained for the European EPHECT project [5, 10] were selected for this HRA.

3.2 Exposure assessment

For each exposure scenario, an AIC_{house} was calculated for each substance emitted by each tested product. Table 3 presents a synthetic overview (minimum, maximum, median and average data) of the obtained results for a selection of substances of interest. Tables S3 and S4 present all of the obtained results for the Common Use scenario and the Reasonable Worst-Case scenario.

Furthermore, an HAC_{max} was calculated for each substance emitted by each tested product. Table 4 presents a synthetic overview of the obtained results for a selection of substances known for their potential acute effects. Table S5 presents all of the obtained results.

3.3 Selection of toxicity values

Table 5 presents the TRVs retained for a selection of substances of interest and the associated critical effects.

No TRV was available for limonene or alpha-pinene in the consulted databases. Consequently, for chronic exposures, the derived EU-LCI values were considered additional information. These EU-LCI values are 2.5 mg/m^3 and 5 mg/m^3, respectively.

3.4 Chronic risk assessment

For each substance emitted by each tested product and under each generic exposure scenario, a TER and an IER were calculated when a TRV_T and a TRV_{NT} were available (Tables S3 and S4). The main obtained results are presented below.

3.4.1 Common Use scenario

For each emitted substance considered individually, no TER and no IER exceeded the usual reference values of 1 and 10^{-5}, respectively. Moreover, for each product, no multisubstance sum of TERs and no multisubstance sum of IERs exceeded the usual reference values. Finally, based on the tested products of the highest risks for each type of product considered in the Common Use session, no multiproduct sum of TERs and no multiproduct sum of IERs exceeded the usual reference values.

3.4.2 Reasonable Worst-Case scenario

For each emitted substance considered individually, no TER and no IER exceeded the usual reference values of 1 and 10^{-5}, respectively. The substances with at least one risk indicator greater than 20% of a usual reference value (i.e., 0.2 or 2.10^{-6}) are four carbonyl compounds: acrolein, crotonaldehyde, propionaldehyde and benzaldehyde.

Exceedances of the multisubstance sum of TERs were obtained for four products, ranging from 1.1 to 1.9. No multisubstance sum of IERs exceeded the usual reference value.

Based on the tested products of greatest risk for each type of product considered in the Reasonably Elevated Use session, the multiproduct sum of TERs reached 4.6 (acrolein is associated with the highest contribution ($\sum$ TERs = 2.7)). The multiproduct sum of the IERs is less than the usual reference value.

3.5 Acute risk assessment

For each substance emitted by each tested product, an ARR was calculated when TRV_{acute} was available (Table S5). The main obtained results are presented below.

3.5.1 Common Use scenario

ARR exceeded the usual reference value of “1” for two substances: (i) acrolein, for five products, with an average ARR of 1.7 and a maximum ARR of 3.3; and (ii) formaldehyde, for three products, with an average ARR of 1.9 and a maximum ARR of 2.4.

3.5.2 Reasonable Worst-Case scenario

ARR exceeded the usual reference value (1) for two substances: (i) acrolein, for eight products, with an average ARR of 1.8 and a maximum ARR of 4.6; and (ii) formaldehyde, for eight products, with an average ARR of 1.5 and a maximum ARR of 3.3.

As additional information, the maximum limonene HAC_{max} (approximately 720 µg/m^3) was far less than the acute toxicity value (90 mg/m^3 – 30-min exposure) established under the European EPHECT project [23].

4 Discussion

4.1 Exposure assessment

This study proposed a method to address the complexity of establishing representative multiroom/multiproduct cleaning sessions. This method aimed to better characterize exposures associated with household cleaning by considering hypotheses closer to the observed uses: in the absence of sufficient data on co-uses, considering a full cleaning session intended to propose an answer to two classic critiques of (regulatory) HRA: (i) quantifying risks product by product, each considered individually, without assessing potential combinations of products, could lead to underestimation of actual risks; and (ii) inversely, totaling of the risk indicators calculated for each product, considered individually, could lead to very high—sometimes unrealistic—aggregate exposures [27, 30, 31, 52] because the corresponding individual exposures are often quantified with (very) conservative hypotheses, especially in a regulatory framework in which the objective is to ensure the absence of health effects.

A probabilistic approach might be another way of characterizing co-uses [27, 51, 52]. This approach presupposes that statistical distributions of uses can be established with a minimum level of uncertainty. The question of their respective correlations could remain a challenge.

The method proposed in this HRA constitutes one type of step further that does not pretend to cover every single case. The objective was to characterize the actual variability based on two points of reference—i.e., a Common Use session and a Reasonably Elevated use session—contextualized with the opinions of stakeholders of various profiles to obtain a more informed view.

Another limitation is inherent to the generic aspect of the considered scenarios: although durations of time could be attributed to each type of cleaning product in the selected rooms, a substantial variety of products can be found within the types of products considered in this study. For example, all-purpose cleaners, floor cleaners and window cleaners are general categories that include multiple subtypes of products, especially regarding formats: liquids, sprays, foams, powders, wipes; for example; the cleaning time associated with wipes is likely much shorter than the cleaning time of other formats [46], and a factor of five could be considered [40].

To our knowledge, this HRA is the first to combine measurements under realistic conditions and an established full cleaning session elaborated with stakeholder participation.

The obtained exposures were calculated from concentrations measured during the two and a half hours following the beginning of use. These field concentrations allowed us to consider the secondary substances formed in the indoor air during the measurement period. This secondary formation could represent a significant contribution to the total measured concentrations, e.g., for formaldehyde [7, 53]. Field concentrations also allowed us to avoid the difficulties of simulating real emissions processes with a laboratory test chamber. These difficulties include controlling various parameters, e.g., humidity rate, oxygen rate, air flow, and temperature [32, 54].

The main limitations associated with the calculation of AICs consisted of the rather simple assumptions chosen to extrapolate the measured concentrations, including a "well-mixed air" approach. This type of hypothesis was used in previous studies [7]. Moreover, household uses are frequently associated with some indirect emissions, e.g., storage-related emissions and emissions during preparation of the product. These emissions were not within the scope of this study.

4.2 Chronic and acute risk characterization

The results obtained for the Common Use scenario suggest that chronic exposures associated with the most common uses are not of concern with regard to the targeted substances, the selected products and the assumptions of this HRA.

Exceedances obtained for the Reasonable Worst-Case scenario were associated with certain multisubstance totals of risk indicators only, and the maximal sum for the full cleaning session was less than 5 (TER). Consequently, considering that this scenario aggregated multiple conservative hypotheses and with regard to the other uncertainties of this HRA, the assessed level of risks can be considered low.

Acrolein and benzene are the substances making the main contributions to the total risks for threshold effects and for nonthreshold effects, respectively. The associated maximum chronic exposures (maximum AIC_{house}), i.e., $7.1.10^{-1}$ µg/m^3 and $2.7.10^{-1}$ µg/m^3, respectively, are less than typical background concentrations that can be found in private dwellings (e.g., acrolein – median in France: 1.1 µg/m^3 [41] and benzene – mean in Europe: 3 µg/m^3 [55]).

For acute exposures, under the Reasonable Worst-Case scenario, four calculated ARRs exceeded 2, ranging from 2.1 to 4.6. The two corresponding products were in wipe format. If the likely hypothesis of overestimation by a factor of 5 (described above) is retained, then no exceedance is actually expected for these two products.

The remaining ARR exceedances less than below 2. The corresponding maximum HAC_{max} values are 12 µg/m^3 for acrolein and 140 µg/m^3 for formaldehyde. When acute exposures exceed the selected TRV_{acute}, the first observed effects are expected to be: (i) nasal and throat irritation and decreased respiratory rate for acrolein; and (ii) eye irritation for formaldehyde; however, if repeated, this irritation could lead to more severe and irreversible effects, such as nasopharyngeal carcinogenic effects.

As additional information, acrolein acute exposures (HAC_{max} up to 12 µg/m^3) can also be compared to the OEHHA Acute Reference Exposure Level (2.5 µg/m^3 - 1 h - respiratory and eye irritation [56]) and to the acute Critical Exposure Limit established for the European EPHECT project (21 µg/m^3 – 30 min - subjective eye irritation [23]).

In conclusion, with regard to the level of uncertainty associated with the hypotheses retained for this HRA, the assessed acute risks can be considered significant, ranging from low to moderate. In particular, the obtained results suggest a need to reduce the emissions of the most emissive products. To this end, several types of public health risk management actions are usually considered, e.g., regulations on composition, regulations on emissions levels, and labeling about emissions levels. The obtained results suggest that these potential actions should prioritize the reduction of acrolein and formaldehyde emissions.

In the scientific literature, few inhalation risk assessments have been conducted for cleaning products, and they considered limited selections of products and targeted substances [24, 57]. These assessments concluded that no situation of concern was expected. This difference from the conclusions of this HRA can be explained by differences in the selected toxicity values and the higher maximum exposures extrapolated from direct emission measurements (e.g., in the literature, no formaldehyde acute exposure exceeds 100 µg/m^3 (max: 82 µg/m^3 for a floor cleaner)).

However, several reviews of the scientific literature, based on epidemiological studies mainly addressing professional uses, have shown an association between the use of cleaning products and various respiratory disorders, including an increased risk of asthma and of the intensity of associated symptoms [16, 17, 20, 21, 58-60]. The strongest level of evidence relates to professional uses, but multiple studies have indicated situations of concern for household use [14, 16, 58, 61]. This HRA suggests that the cumulative risks attributable to substances emitted by cleaning products might exceed the usual reference values, but the exceedances are mainly of small magnitudes while corresponding to a scenario integrating multiple reasonably conservative hypotheses. However, given the current state of knowledge and considering their construction method, TRVs imperfectly protect against respiratory sensitizing effects.

4.3 Priority substances of interest

Based on all of the calculated inhalation risk indicators of this HRA, the identified substances of greater interest are acrolein, formaldehyde and, to a lesser extent, benzene, crotonaldehyde, propionaldehyde and benzaldehyde.

These substances of interest are not specific to the emissions from cleaning products, especially acrolein, formaldehyde and benzene; multiple other sources of emissions are commonly present in indoor environments, e.g., furniture, air fresheners, tobacco smoke, incense, construction products, paints, cooking activities, and scented candles. Consequently, cumulative exposures are expected and could lead to higher risks than those identified for each source, considered individually. Background concentrations in European private dwellings are available for formaldehyde (minimum: 7 µg/m^3; maximum: 57 µg/m^3; mean: 22 µg/m^3) and benzene (minimum: 0 µg/m^3; maximum: 32 µg/m^3; mean: 3 µg/m^3) [10, 55, 62]. Acrolein background concentrations are available for French private dwellings (95th percentile: 3.4 µg/m^3; median: 1.1 µg/m^3) [41]. The health reference values considered in this HRA relate to total indoor air concentrations; in particular, they do not relate to the concentrations attributable to the use of cleaning products only. This fact suggests a need to limit the emissions of cleaning products, especially for the identified priority substances of interest, until typical cumulative exposures are less than the selected health values.

Furthermore, household uses of cleaning products can also produce fine particulate matter ($PM_{2.5}$) in indoor air, including secondary particles, e.g., particles produced by ozone-induced chemistry in the presence of terpenes [9, 39, 63]. Some of these particles $PM_{2.5}$ can be submicron (PM_1) in size; and some are ultrafine ($PM_{0.1}$) [24, 63, 64].

For the products tested in this study, particles with a size of approximately 50 nm could reach 160,000 particles/cm^3. Compared to background levels of approximately 4,000 particles/cm^3, this increase can be considered a point of vigilance that should be further studied from a health point of view: in general, submicron and ultrafine particles are subjects of concern because of their higher surface reactivity and their ability to penetrate the pulmonary system [65, 66].

4.4 Risk perception

Household uses of cleaning products emit substances of concern in indoor air. However, because some users assume that infectious diseases are caused by the presence of microorganisms, seeking a high level of cleanliness can be part of a preventive logic, especially in the presence of children [67, 68]. This preventive logic could be reinforced by marketing and advertising strategies, especially during the global COVID-19 pandemic. For example, in online marketplaces, cleaning products could be associated with the following advertisements: "*5-in-1 virucidal cleanser",* "*Anti-virus*", *"Cleans, disinfects, removes all mold"*, *"eliminates and prevents mold development", "cleanse your surfaces", "destroys 99.9% of bacteria, fungi and viruses*", and "*compliant with European antimicrobial efficacy standards".*

This type of perception could increase the level of cleaning product use. Therefore, it is not surprising that 15 % of all-purpose cleaning product users in Europe use more products than recommended by the manufacturer [4].

5 Conclusions

The health risk assessment (HRA) approach was applied to indoor air emissions from nineteen cleaning products tested under realistic conditions (under typical indoor environmental parameters) in an experimental house.

Full cleaning sessions were elaborated with data available in the technical and scientific literature, combined with stakeholder participation (e.g., manufacturers, professional and private housekeepers, environmental health associations, consumer organizations). These sessions included durations of use for different types of products in each typical room of a private dwelling (e.g., kitchen, bathroom, living/dining room, bedrooms).

Two generic exposure scenarios were elaborated: a Common Use scenario, aiming to characterize a user whose practices and environmental characteristics (volume of the rooms, air change rate, etc.) are common, being approximately the average of the observed practices and characteristics; and a Reasonable Worst-Case scenario, aiming to characterize a user whose practices and environmental characteristics reasonably increase the average exposure. The Common Use scenario included a one-and-a-half-hour cleaning session once per week, followed by manual ventilation; the Reasonable Worst-Case Scenario included a four-hour session twice per week, with no manual ventilation.

To our knowledge, this HRA is the first to combine concentrations measured under realistic conditions with full cleaning sessions.

The obtained results for the Common Use scenario suggest that chronic exposures associated with the most common uses are not of concern with regard to the targeted substances, the selected products and the assumptions of this HRA.

The obtained results for the Reasonable Worst-Case scenario suggest that the highest chronic exposures are associated with low risks. In particular, no risk indicator exceeded the selected usual reference values for a substance considered individually; only multisubstance/multiproduct sums of indicators exceeded the selected health reference values.

However, the obtained results for acute (1-hour) exposures suggest a need to reduce emissions from the most emissive products, especially for acrolein (up to 12 µg/m^3) and formaldehyde (up to approximately 140 µg/m^3).

Acrolein and formaldehyde are not specific to cleaning product emissions: cumulative exposures are expected with other household products, e.g., paints, incense, scented candles, furniture, and fragrance diffusers.

Emitted $PM_{2.5}$ particles could also be of concern because a large number of them are submicron and ultrafine (higher surface reactivity and greater ability to penetrate the pulmonary system).

The obtained results provide a complementary perspective to the results of the European EPHECT project [10, 23]: this HRA focused on cleaning products based on actual indoor air concentrations (experimental values instead of modeled concentrations) and considered a large set of substances. The same strategy, combining experiments under realistic conditions and stakeholder input regarding uses, could be implemented to assess the health risks of many other usual consumer products. The scope of these potential future assessments could also be extended to SVOCs.

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Conflicts of Interest

The authors declare that they have no conflicts of interest.

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Credit Authorship Contribution Statement

Guillaume KARR: Conceptualization, Formal Analysis, Methodology, Writing – Original Draft; Mélanie NICOLAS: Resources, Investigation, Validation, Data Curation, Writing – Review and Editing; François MAUPETIT: Writing – Review and Editing, Project Administration; Martine RAMEL: Conceptualization, Methodology, Validation, Writing – Review and Editing, Funding Acquisition, Project Administration

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Table 1: Cleaning sessions selected for the health risk assessment (HRA)

Common Use Session	Reasonably Elevated Use Session

Once a day (average durations of use):
 	
• Kitchen
  ○ Dishwashing product: 10 min
  ○ All-purpose cleaner: 10 min
 	
In addition, once a week (average durations of use):
 	
• Kitchen
  ○ All-purpose cleaner: 15 min
  ○ Floor cleaner: 10 min
  ○ Window cleaner: 10 min (once every 3 weeks)
• Bedrooms (2) – time per room
  ○ Floor cleaner: 5 min
  ○ Window cleaner: 5 min (once every 3 weeks)
• Bathroom
  ○ Bathroom cleaner: 10 min
  ○ Bleach to dilute: 5 min
  ○ Floor cleaner: 5 min
  ○ Window cleaner: 5 min (once every 3 weeks)
• Toilet
  ○ Floor cleaner: 5 min
  ○ WC cleaner: 5 min
• Living/dining room
  ○ Floor cleaner: 15 min
  ○ Window cleaner: 10 min (once every 3 weeks)
	Once a day (average durations of use):
	
	• Kitchen
	  ○ Dishwashing product: 30 min
	  ○ All-purpose cleaner: 20 min
	  ○ Floor cleaner: 5 min
	• Toilet
	  ○ WC cleaner: 5 min
	• Bathroom
	  ○ Bathroom cleaner: 10 min
	
	In addition, twice a week (average durations of use):
	
	• Kitchen
	  ○ All-purpose cleaner: 20 min
	  ○ Floor cleaner: 10 min
	  ○ Window cleaner: 10 min
	• Bedrooms (4) – time per room
	  ○ Dust cleaner: 10 min
	  ○ Floor cleaner: 10 min
	  ○ Window cleaner: 5 min
	• Bathroom
	  ○ Bathroom cleaner: 15 min
	  ○ Bleach to dilute: 10 min
	  ○ Floor cleaner: 10 min
	  ○ Window cleaner: 5 min
	• Toilet
	  ○ WC cleaner: 10 min
	  ○ Bleach to dilute: 5 min
	• Living/dining room
	  ○ Floor cleaner: 20 min
	  ○ Dust cleaner: 15 min
	  ○ Window cleaner: 10 min
	• Cellar/storage room:
	  ○ All-purpose cleaner: 5 min

Weekly total: 1 session per week, lasting 1 hour and 35 minutes	Weekly total: 2 sessions per week, lasting 4 hours and 5 minutes each

Table 2: Generic exposure scenarios elaborated for the health risk assessment

Scenario components		Cleaning product users	
		Reasonable Worst Case	**Common Use**
Frequencies and durations of use (/day)	Restroom (WC)	Reasonably Elevated Use Session	Common Use Session
	Living/dining room		
	Kitchen		
	Entrance hall		
	Bathroom		
	Bedroom(s)		
	Cellar/storeroom		
Presence in the room during and after use		Yes	Yes
Years of exposure (years)		70	70
Manual ventilation (opening to the outside)		No manual ventilation	10 min after use
Air change rate (/h)		0.35	0.35

Table 3: Chronic exposures for a selection of substances of interest – average inhaled concentrations (AIC_{house}, µg/m^3)

Substances	**CAS number**	**Common Use Scenario**				**Reasonable Worst-Case Scenario**			
		Min.	**Max.**	**Med.**	**Avg.**	**Min.**	**Max.**	**Med.**	**Avg.**
Formaldehyde	50-00-0	0.0	1.0×10^{-1}	3.3×10^{-3}	2.1×10^{-2}	0.0	1.3×10^{1}	1.0	2.3
Benzene	71-43-2	0.0	1.9×10^{-2}	0.0	1.2×10^{-3}	0.0	2.7×10^{-1}	6.9×10^{-3}	4.2×10^{-2}
Acetaldehyde	75-07-0	0.0	1.2×10^{-1}	5.9×10^{-4}	1.2×10^{-2}	0.0	4.7	2.3×10^{-1}	7.1×10^{-1}
D-Limonene	5989-27-5	0.0	1.8	2.3×10^{-2}	2.5×10^{-1}	0.0	3.7×10^{1}	8.8×10^{-1}	5.4
Acrolein	107-02-8	0.0	2.5×10^{-2}	3.9×10^{-5}	2.1×10^{-3}	0.0	7.1×10^{-1}	3.8×10^{-2}	1.5×10^{-1}
Acetonitrile	75-05-8	0.0	6.9×10^{-2}	2.3×10^{-5}	1.7×10^{-2}	0.0	9.6	9.0×10^{-2}	2.4
Acetone	67-64-1	0.0	2.3×10^{1}	1.1×10^{-3}	7.7×10^{-1}	0.0	2.0×10^{3}	4.6×10^{-1}	7.3×10^{1}
Acetic acid	64-19-7	0.0	9.3×10^{-3}	2.9×10^{-5}	3.1×10^{-3}	3.9×10^{-3}	8.1	7.6×10^{-2}	2.7
Isopropanol	**67-63-0**	0.0	3.2	7.4×10^{-3}	3.5×10^{-1}	0.0	7.7×10^{1}	5.6	2.0×10^{1}

Note: Min.: minimum; Max.: maximum; Med.: median; Avg.: average

Table 4: Calculated acute exposures for a selection of substances of interest – maximum 1-hour average concentrations (HAC_{max}, µg/m³)

Substances	CAS Number	Common Use Scenario				Reasonable Worst-Case Scenario			
		Min.	Max.	Med.	Avg.	Min.	Max.	Med.	Avg.
Formaldehyde	50-00-0	0.0	2.4×10^{2}	2.6×10^{1}	4.5×10^{1}	0.0	3.3×10^{2}	5.4×10^{1}	6.7×10^{1}
Acetaldehyde	75-07-0	0.0	1.0×10^{2}	8.3	1.5×10^{1}	0.0	4.7×10^{2}	1.3×10^{1}	4.5×10^{1}
Acrolein	107-02-8	0.0	2.3×10^{1}	2.4	3.8	0.0	3.2×10^{1}	3.6	5.6
Acetone	67-64-1	0.0	2.3×10^{4}	2.2×10^{1}	1.2×10^{3}	0.0	3.0×10^{4}	4.4×10^{1}	1.7×10^{3}
Benzene	71-43-2	0.0	2.1×10^{1}	6.1×10^{-1}	2.6	0.0	3.2×10^{1}	8.2×10^{-1}	4.9

Note: Min.: minimum; Max.: maximum; Med.: median; Avg.: average

Table 5: Values characterizing the toxicity of several substances of interest

CAS number	Substances	TRV_T (µg/m³)	Org.	Date	TRV_{NT} (µg/m³)	Org.	Dated	TRV_{acute} (µg/m³)	Org.	Date	Critical effects
50-00-0	Formaldehyde	-	-	-	-	-	-	1.0×10^{2}	WHO	2010	Acute: Subjective and objective eye irritation
75-07-0	Acetaldehyde	1.6×10^{2}	ANSES	2014	-	-	-	3.0×10^{3}	ANSES	2014	Threshold: Degeneration of the olfactory epithelium Acute: Bronchoconstriction in asthmatic patients
107-02-8	Acrolein	1.5×10^{-1}	ANSES	2019	-	-	-	6.9	ATSDR	2007	Threshold: Lesions of the upper respiratory epithelium Acute: Nasal and throat irritation, decreased respiratory rate
108-88-3	Toluene	2.0×10^{4}	ANSES	2018	-	-	-	2.0×10^{4}	ANSES	2018	Threshold: Neurological effects (color vision disorders) Acute: Neurological effects
91-20-3	Naphthalene	1×10^{1}	WHO	2013	5.6×10^{-6}	ANSES	2013	-	-	-	Threshold: Lesions of the respiratory and olfactory epithelium Non-Threshold: Neuroblastomas of the olfactory epithelium
67-64-1	Acetone	3.3×10^{4}	ATSDR	1994	-	-	-	6.6×10^{4}	ATSDR	1994	Threshold: Neurological effects Acute: Neurological effects
71-43-2	Benzene	1.0×10^{1}	ANSES	2010	2.6×10^{-5}	ANSES	2013	2.7×10^{1}	OEHHA	2014	Threshold: Immunological disorders Non-Threshold: acute leukemia Acute: reproductive disorders, aplastic anemia and acute myeloid leukemia
67-63-0	Isopropanol	7.0×10^{3}	OEHHA	2000	-	-	-	-	-	-	Threshold: Kidney lesions in mice and rats; fetal growth retardation and developmental anomalies in rats

Notes:

- † *TRV_T stands for threshold toxicological reference value for the inhalation route and for chronic exposure;*
- † *TRV_{NT} stands for no-threshold toxicological reference value for the inhalation route and for chronic exposure;*
- † *TRV_{acute} stands for acute toxicological reference value for the inhalation route;*
- † *Date means date of construction or date of last revision;*
- † *Org. means producing organization;*
- † *for formaldehyde, the absence of chronic effects is ensured by compliance with the TRV_{acute};*
- † *for acetaldehyde, the absence of carcinogenic effects is ensured by compliance with TRV_T.*